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(a) time-resolved infrared spectral photography
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(c) parahydrogen 16μm tunable source
(d) $c^3\Pi_u$ state of H_2

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Results achieved in four separate laser studies are summarized. Topics covered are (a) time-resolved infrared spectral photography, (b) laser-induced thermal explosions, (c) parahydrogen 16μm tunable source, and (d) a study of the $c^3\Pi_u$ state of H_2 in discharges.

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Study of Lasers

Final Report

Peter P. Sorokin

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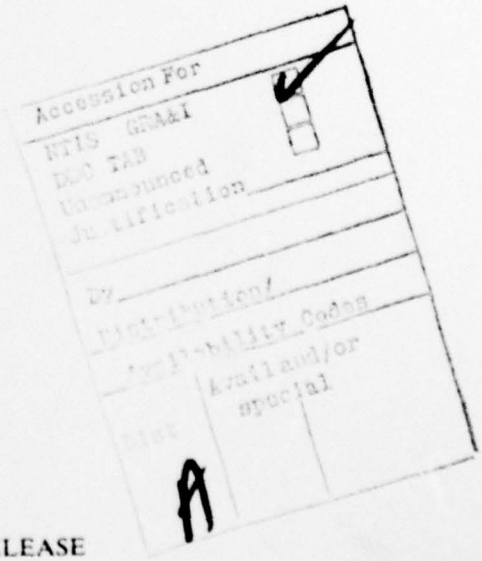

I. Introduction

The work of the contract just concluded comprises several studies. In rough order of importance, the topics are as follows: 1) time-resolved infrared spectral photography, 2) laser-induced thermal explosions, 3) 16μm tunable i.r. source utilizing four-wave mixing in parahydrogen, and 4) characterization of the $c^3\Pi_u$ state of H_2, with a view towards two-photon amplification. Results achieved are briefly described below.

II. Principal Results

(a) Time-Resolved Infrared Spectral Photography

During the contract period a new technique was developed[1,2] which allows broadband infrared absorption spectra to be photographed with ≈5nsec time resolution, even under some conditions, in a single-shot. Experimental details and theoretical aspects of this method are discussed at length in [2]. Briefly, the technique utilizes third order nonlinearities of alkali metal vapors to achieve two objectives: (1) generation of a pulsed, broadband, infrared continuum beam (ν_{ir}) which can probe the absorption of a sample and (2) subsequent conversion of this i.r. beam, with its encoded spectral information about the sample, to the visible, where it can be photographically or photoelectrically recorded. The two principal nonlinear processes involved are diagrammed in Fig. 1. Generation of ν_{ir} is accomplished by broadband stimulated electronic Raman scattering (SERS) in a first alkali metal vapor heat-pipe oven (HP1 in Fig. 2). A broadband ($\Delta\nu$~1000cm^{-1}) visible dye laser beam ν_c originates from a mirrorless, superfluorescent dye cell (C_1 in Fig. 2) and is subsequently amplified to the level of a few millijoules and focused into HP1. This visible beam, the primary beam for the SERS process, is converted to an infrared Stokes beam having approximately the same spectral width and pulse duration (≈5nsec) as ν_c. The i.r. spectral range spanned by ν_{ir} depends on both ν_c and the energy of the final electronic state involved in the Raman transition. For Rb, with ν_c in the vicinity of the 6p resonance lines, the infrared

generated falls in the 2.7μm range. With the use of dye laser continua spectrally located near other alkali resonance lines, generation of broadband i.r. in several different regions, extending out to at least 30μm, should be possible.[1]

The second nonlinear process diagrammed in Fig. 1 is responsible for the upconversion that occurs in the second heat-pipe oven (HP2 in Fig. 2). A narrow band laser beam ν_L, injected into HP2 via a silicon wafer (Si_2), generates by SERS a narrow-band Stokes beam ν_S. Beams ν_L, ν_S and ν_{ir} beat together in HP2 to produce an upconverted beam ν_U by means of the resonantly enhanced four-wave mixing process shown in Fig. 1. The intensity of a given spectral component of ν_U is linearly proportional to its corresponding ν_{ir} component. Thus infrared spectra are simply translated to the visible by the fixed Raman energies of the alkali metal atoms in HP2. This procedure allows one to take advantage of the fact that detectors

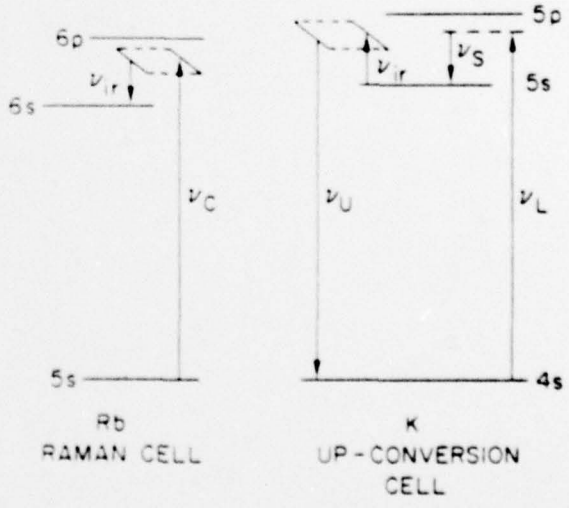

Figure 1 Level diagrams for the Raman generation and four-wave upconversion processes.

with high inherent spectral resolution exist for the visible region.

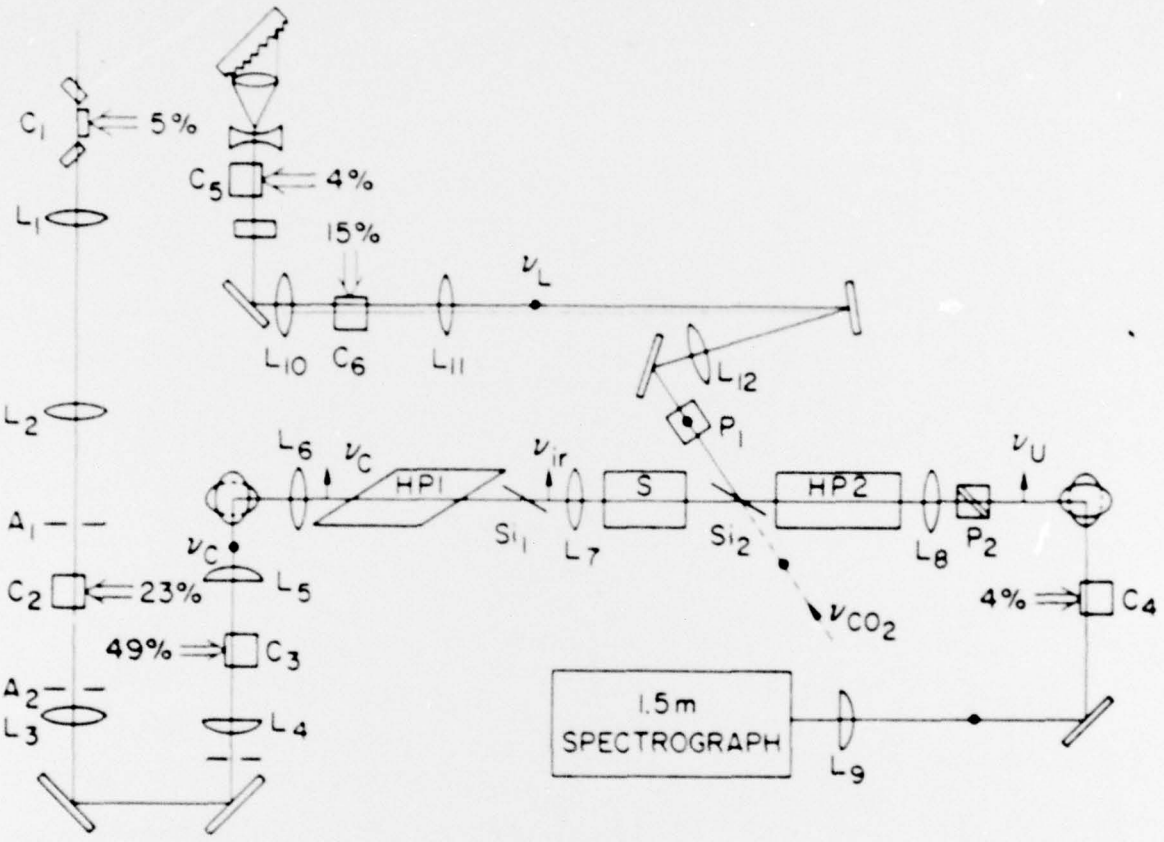

Figure 2 Diagram of experimental apparatus.

Infrared spectra of several molecules (CO_2, H_2O, NH_3, CH_4, CH_3NC, tetrahydropyran quadricyclane) were recorded with this technique.[2] Transient spectra of the laser induced isomerization $CH_3NC \rightarrow CH_3CN$ were also recorded.[2] With [illegible] vapor used in HP1, the continuous range 2500-4000cm^{-1} has now been probed.

Recent efforts in our laboratories have shown that dye cells C_1, C_2, and C_3 in Fig. 2 can effectively be replaced with a single high output broadband dye laser leading to simplification

of the apparatus. Also it has been demonstrated that the Nd^{3+}:YAG pump laser originally used[1,2] can effectively be replaced with a XeCl excimer laser. This potentially allows i.r. probing to ~30μm.

A patent application on this technique was recently filed by IBM.[3]

(b) Laser-Induced Thermal Explosions (LITE)

Application of single pulses of CO_2 TEA laser radiation to a vapor of methyl isocyanide (CH_3NC), with the laser tuned to coincide with the fundamental frequency ν_4 of this molecule, resulted in efficient conversion of the gas to its isomer, methyl cyanide (CH_3CN).[4] The experiments, performed at room temperature at pressures 10-100 Torr, were interpreted in terms of laser induced heating which is sufficient to trigger a thermal explosion.[4] Time-resolved spectra of this reaction were recorded.

It should be emphasized that LITE is an effect generally realizable with exothermic reactions. The initiating CO_2 or HF infrared laser beam need not necessarily be focused in the vapor. It is only necessary that the following threshold condition[4] be satisfied:

$$\frac{2\kappa}{r_0^2 \epsilon n A} = \frac{e^{-E/R(T_0+\Delta T)}}{\Delta T} \qquad (1)$$

Here ΔT is the minimum initial temperature rise needed for the thermal explosion to develop. The other parameters appearing in Eq. (1) are as follows:

κ	thermal conductivity of gas
n	number density of reacting molecules
r_0	radius of laser beam in gas
ϵ	heat of reaction
A	Arrhenius pre-exponential factor
E	Arrhenius activation energy

With an unfocused infrared laser beam the reaction can be uniformly initiated over large volumes. It consequently can be made to develop in relatively short times, without the occurrence of slow propagation effects. We have recently observed laser induced thermal explosions occurring on a microsecond time scale.

Other examples of LITE reactions, currently under investigation in our laboratory, are the unimolecular decomposition of deutero-hydrazoic acid, and the unimolecular isomerization of dewar hexafluorobenzene (DHFB) to its valence isomer, hexafluorobenzene (HFB). Both reactions are suitable for study by time-resolved spectral photography.

(c) 16μm Tunable I.R. Source Utilizing Four-Wave Mixing in Parahydrogen

A 16μm radiation source based upon the combined effects of stimulated rotational Raman scattering and resonantly enhanced four-wave mixing in parahydrogen gas was demonstrated.[5,6] The maximum output in [6] was measured to be ~40μJ/pulse, which was near the theoretical limit for the 2.5MW CO_2 laser intensities that were applied. Subsequently, in a patent application[7] filed by IBM, it was shown that if an HF or DF laser was used as the Raman driver, in place of the visible laser originally used in the scheme, outputs of ~40mJ at ~16μm could be expected with a ~1J CO_2 laser. This scheme also provides an output that can be easily fine tuned, since there is no threshold requirement involving the CO_2 laser, and the latter may thus be chosen to be a high pressure, continuously tunable laser.

(d) Studies of the $c^3\Pi_u$ State of H_2

Significant populations in the metastable $c^3\Pi_u$ manifold of H_2 were obtained in a pulsed discharge. This manifold potentially could serve as the upper state for two-photon stimulated emission to the autodissociating $b^3\Sigma_u^+$ level. To evaluate this possibility, studies of the population densities and quenching times of some of these levels were made using pulsed laser techniques. A peak density of $10^{12}/cm^3$ in the $c^3\Pi_u$ manifold was measured, with observed quenching times of approximately 10nsec. Similar densities were observed in the $a^3\Sigma_g^+$

manifold. This state could potentially be utilized to produce one-photon stimulated emissions a→b, tunable from 2 to 7eV. The spectral dependences of both one- and two-photon cross sections were calculated. It was found that the two-photon cross section c→b peaks at 1.3eV with a value of $\sim 10^{-52} cm^4$ sec while the one-photon cross section a→b peaks at 3eV with a value $\sim 3 \times 10^{-18} cm^2$. In view of the relatively small cross section for two-photon emission in this case, it is clear that relatively large ($\sim 10^{15}/cm^3$) excited state densities are required. The one-photon tunable UV laser is more favorable, with densities only $\sim 10^{14}/cm^3$ needed.[8]

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